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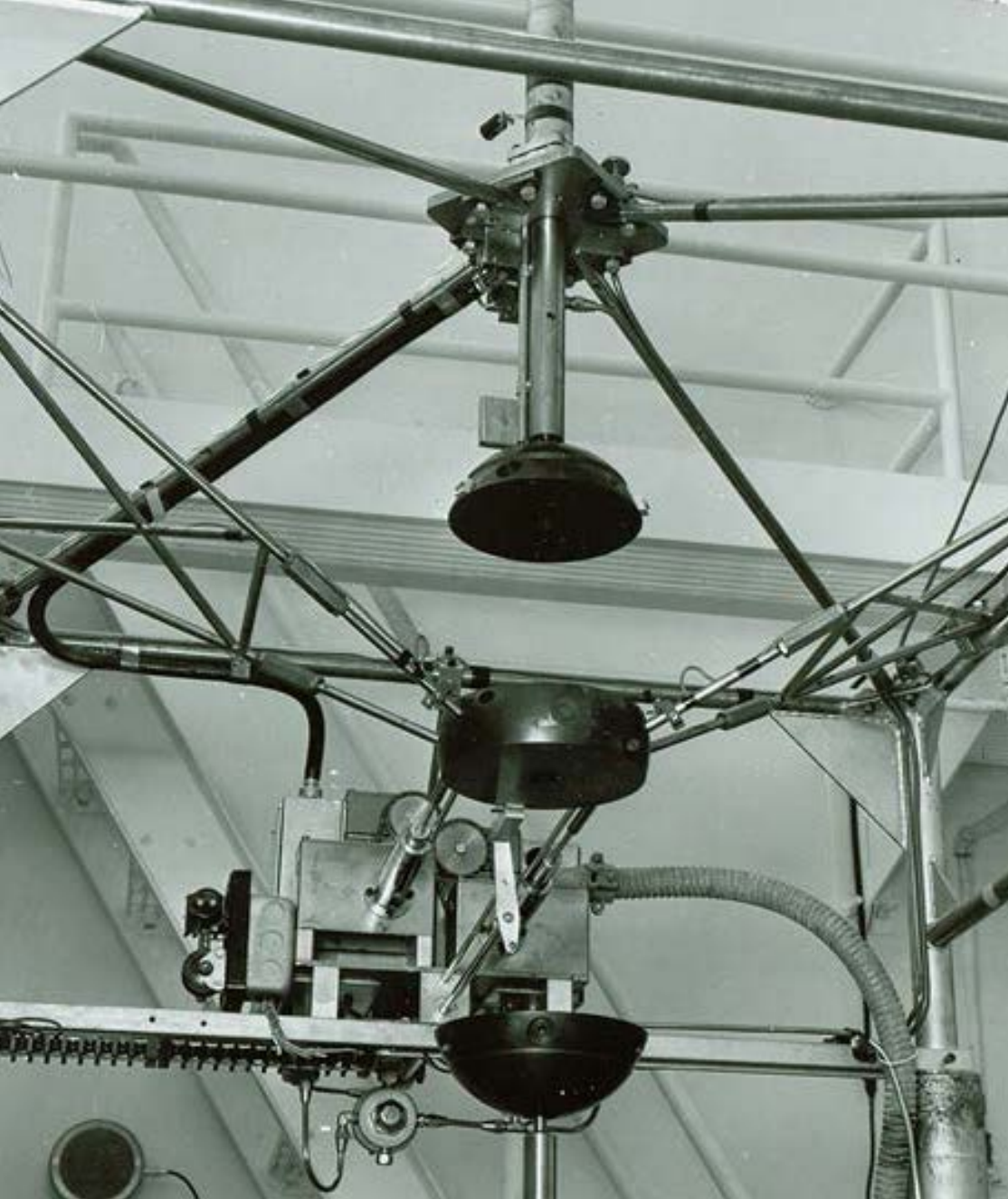
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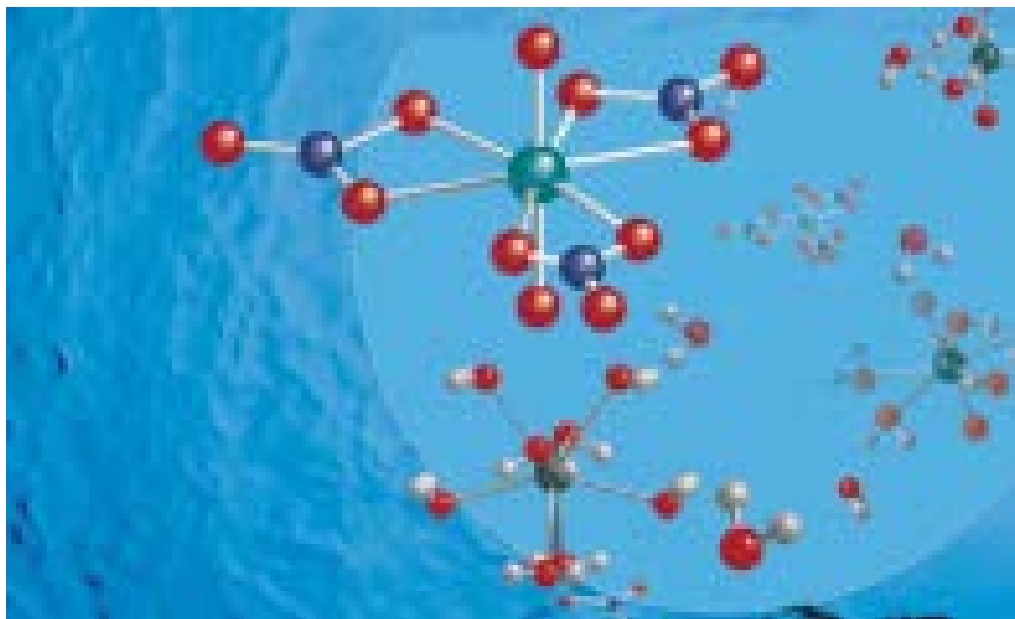
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Experimental Design to Study Criticality Effects of Plutonium Aging

IER-301



**Theresa Cutler, Travis Grove,
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NCSP Technical Program Review
March 2017

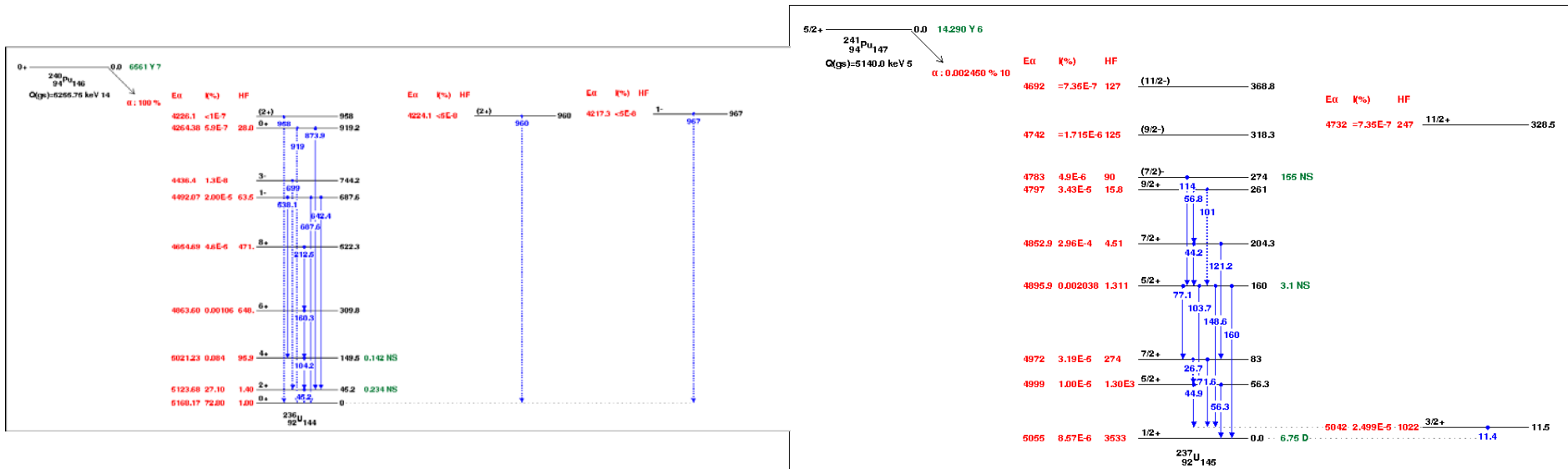
Overview

- **Experimental Objectives**
- **Background and Previous Work**
- **Preliminary Design**
- **Final Design**



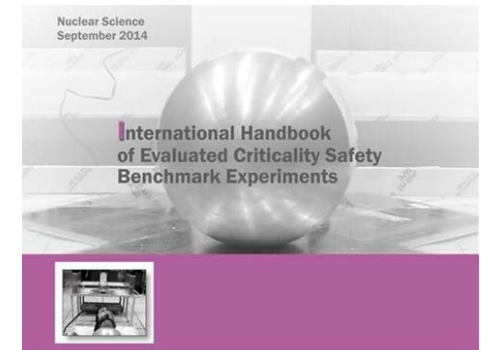
Experiment Objectives

- Examine the effects of plutonium aging on criticality
 - It is known that the isotopic composition of plutonium changes with age, due to radioactive decay
 - Pu-241 \rightarrow Am-241
 - He-3 ingrowth
- The effects of criticality have yet to be examined in a systematic approach
- NEED integral data



Previous Work

- **Metallurgical Effects have been investigated over the past few decades**
 - Much information has been obtained
- **He-3 atoms formed from alpha decay**
 - These atoms migrate toward each other forming sub-micron-size bubbles
 - Become effective “hardening agents” in the plutonium
 - Well documented in the last two decades
- **Accounted for in benchmark evaluations**
 - ZPPR critical benchmark experiments (1969→1980)
 - BeRP ball fundamental physics benchmark experiments (1980→2009)



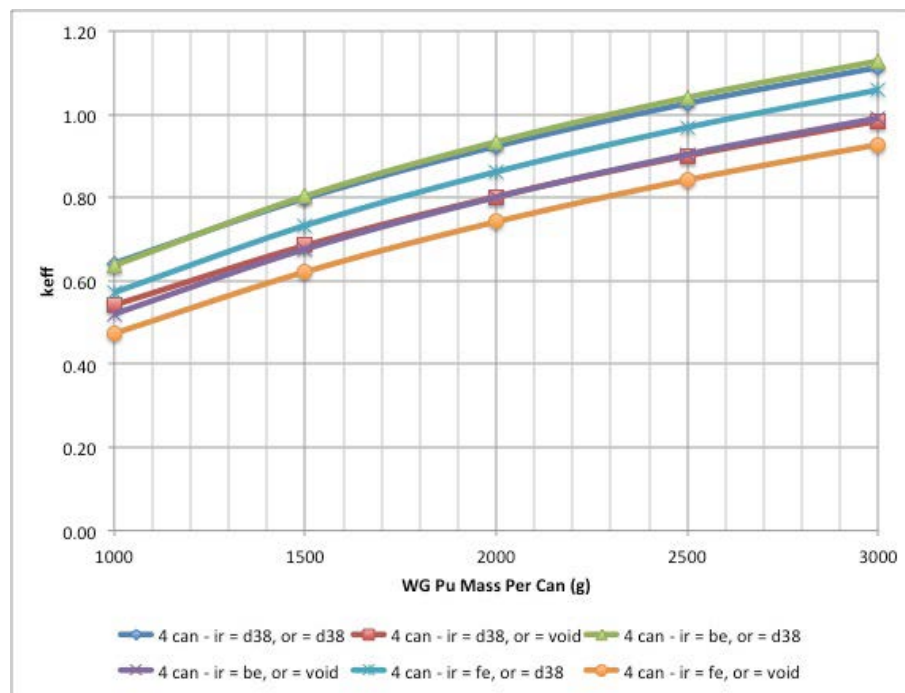
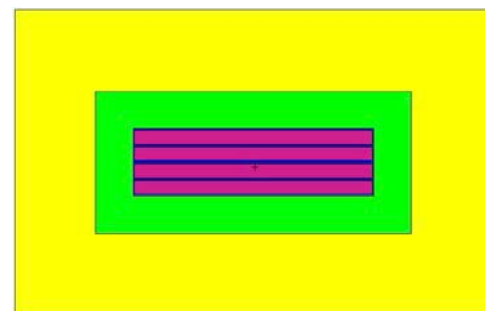
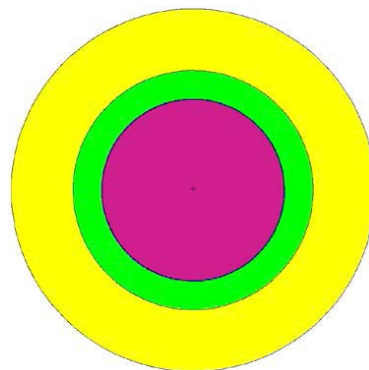
Preliminary Design

- **Design an integral experiment to compare new and aged plutonium**
- **Two approaches considered**
 - **Approach 1:** Design and build a completely new set of critical experiment parts to perform the comparison
 - **Approach 2:** Utilize currently available critical parts to perform small sample reactivity worth comparisons
 - Both options utilize the existing critical assemblies at NCERC



Preliminary Design: Approach 1

- Manufacture 6" OD discs, matching current NCERC inventory and available reflectors (Be, DU, Fe, ...)
- 2 sets
 - One with "new" isotopics
 - One with "aged" isotopics
- All cladding would be of the same material type and thickness



Preliminary Design: Approach 2

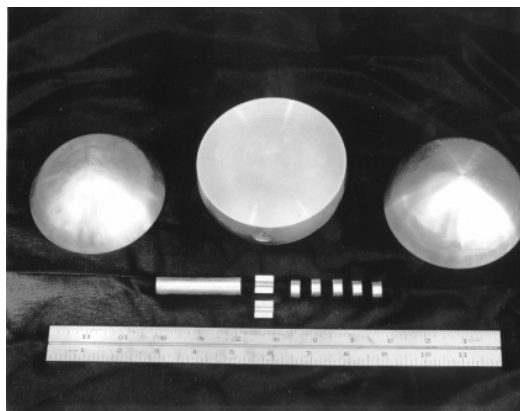
- Utilize currently available critical parts to perform small sample reactivity worth comparisons
 - Flattop glory hole pieces
 - BeRP ball
 - Thor core pieces
- Used by R. Sanchez* to estimate the critical mass of Np-237 using Flattop



BeRP ball



Flattop

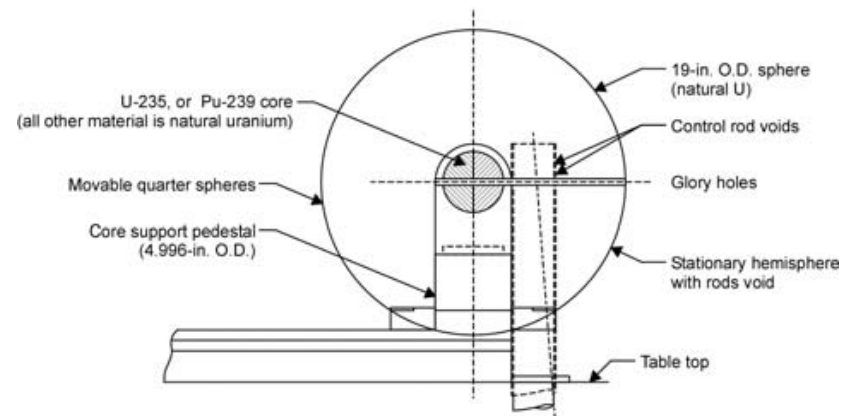


Thor

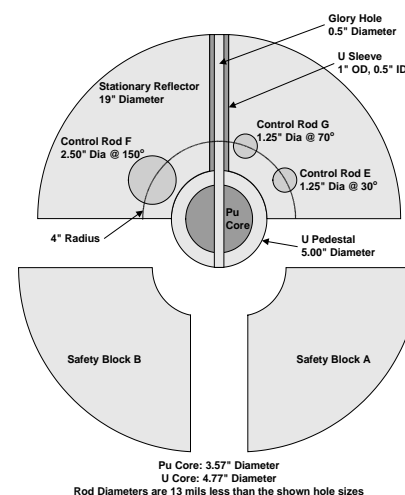
*SPEC-MET-FAST-003, Neptunium-237 and Highly Enriched Uranium Replacement Measurements Performed Using Flattop, 1999-09-30.

Final Design

- **Small Sample glory hole replacement measurements in Flattop**
 - Chosen through systematic evaluation in MCNP and feasibility
 - Future work will likely build upon this with a new full Flattop Pu core
- **Use similar protocol for determining reactivity worth and critical mass to Sanchez Np-237 work in 1990s using the Flattop core**
- **Considered both HEU and Pu core**
 - Pu core selected due to higher worth of glory hole pieces

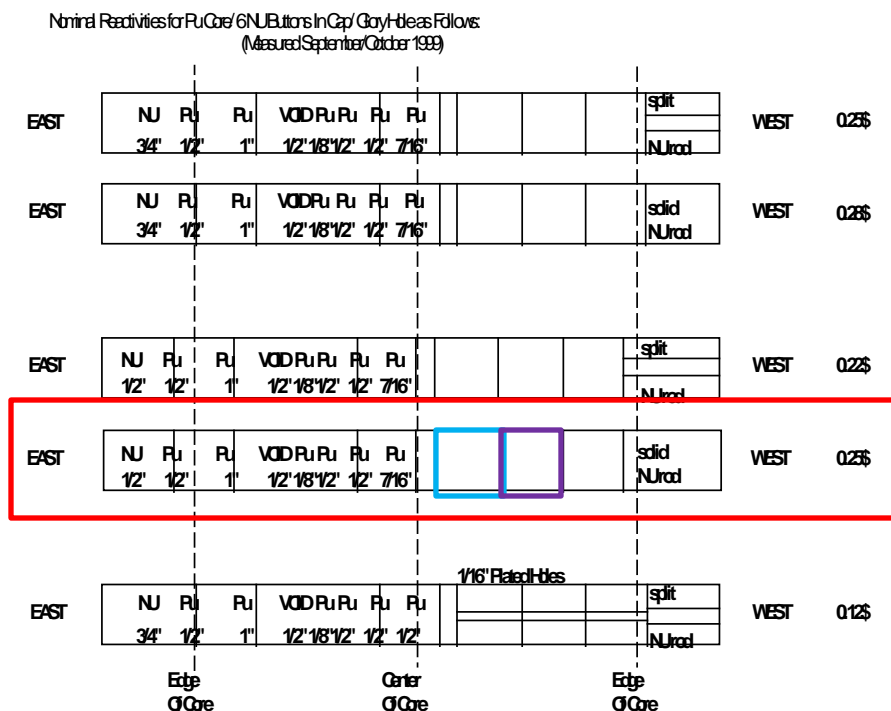


Elevation View



Final Design: Overview

- Glory hole loading with “new” and “old” Pu
- The Flattop Pu core and glory hole pieces were manufactured in ~1957 at Los Alamos
- Diameter of Flattop Pu core glory hole is 0.5”. All pieces used fit tightly
- New pieces, 0.5” OD x 0.5” L, will be manufactured at Los Alamos
- New pieces will be included in the configuration and compared to reactivity results from old configuration
- Locations selected based on highest reactivity worth



Glory Hole Loadings and Associated Reactivity for Flat-top Pu core

Final Design: Isotopics

- **Manufactured in ~1957, δ -phase plutonium, stabilized in gallium**
- **Approximate density: 15.83 g/cm³**
- **5 mil thick nickel cladding on all pieces**
 - New pieces will have 10 mil thick Invar cladding
- **Computationally decayed, using MISC, to 2016**
- **No burnup assumed**
- **No density change effects considered in decay, although acknowledged it decreased slightly from He-3 ingrowth**

Initial Isotopic Composition

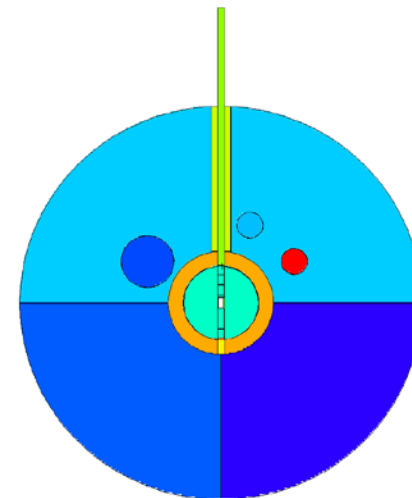
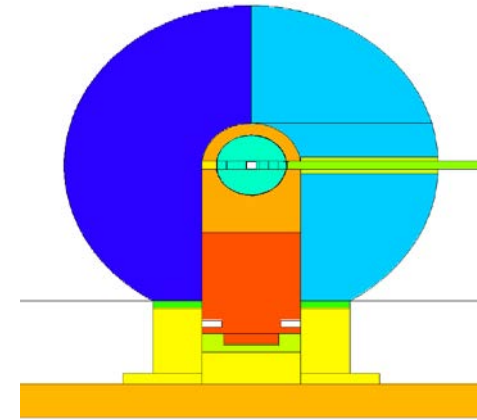
Nuclide	Weight Percent
²³⁹ Pu	93.8
²⁴⁰ Pu	4.8
²⁴¹ Pu	0.3
⁶⁹ Ga	0.6611
⁷¹ Ga	0.4389

Approximate 2016 Decayed Isotopic Composition

Nuclide	Weight Fraction
⁶⁹ Ga	6.611E-03
⁷¹ Ga	4.389E-03
²⁰⁷ Pb	4.889E-15
²²⁷ Ac	1.105E-14
²²⁹ Th	8.384E-14
²³¹ Th	6.140E-15
²³² Th	2.352E-10
²³¹ Pa	4.165E-11
²³³ Pa	5.807E-12
²³³ U	1.237E-09
²³⁵ U	1.510E-03
²³⁶ U	2.834E-04
²³⁷ U	5.895E-12
²³⁷ Np	1.719E-04
²³⁹ Pu	9.365E-01
²⁴⁰ Pu	4.771E-02
²⁴¹ Pu	1.890E-04
²⁴¹ Am	2.636E-03

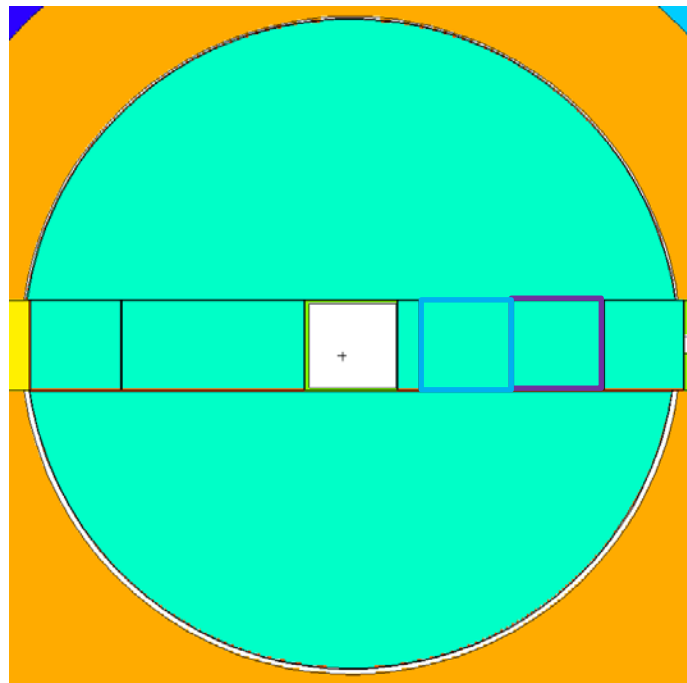
Final Design: MCNP®6 Simulations

- **Detailed Flattop model for HEU core, adapted to the Pu core**
- **Pu core based on detailed engineering drawings**
 - Different than those used in previous benchmarks with Flattop
- **All blues, orange, and reds represent natural uranium; teal represents plutonium.**



Final Design: MCNP®6 Simulations

- Close up view of Pu core with the proposed glory hole loading
- Blue and purple boxes represent the pieces that will be replaced with new ones
- All blues, orange, and reds represent natural uranium; teal represents plutonium.
- HEU and blank pieces also considered in the same locations



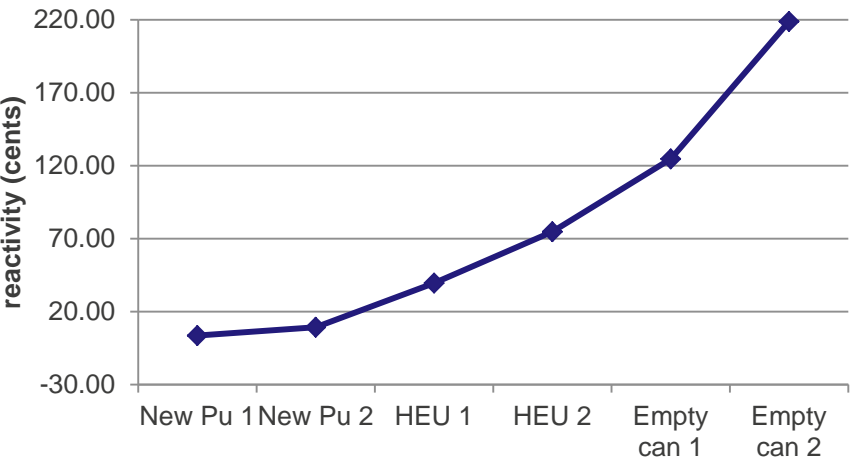
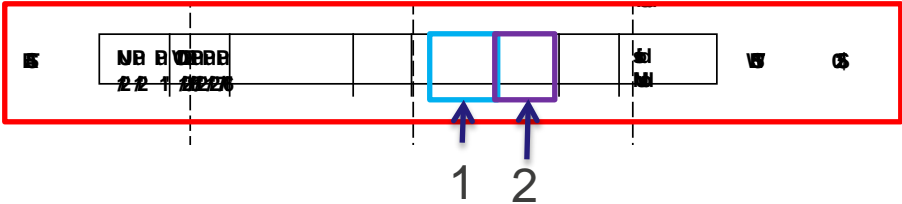
Final Design: MCNP®6 Simulations

MCNP6 keff and Reactivity Results for Expected Replacement Measurements

configuration	keff	delta_keff	reactivity (cents)	delta reactivity (cents)
Base (all old Pu)	1.00269	0	96.85	
New Pu 1	1.00259	0.00010	93.26	3.59
New Pu 2	1.00243	0.00026	87.51	9.34
HEU 1	1.00159	0.00110	57.31	39.54
HEU 2	1.00061	0.00208	22.01	74.84
Empty can 1	0.99923	0.00346	-27.82	124.67
Empty can 2	0.99663	0.00606	-122.07	218.92

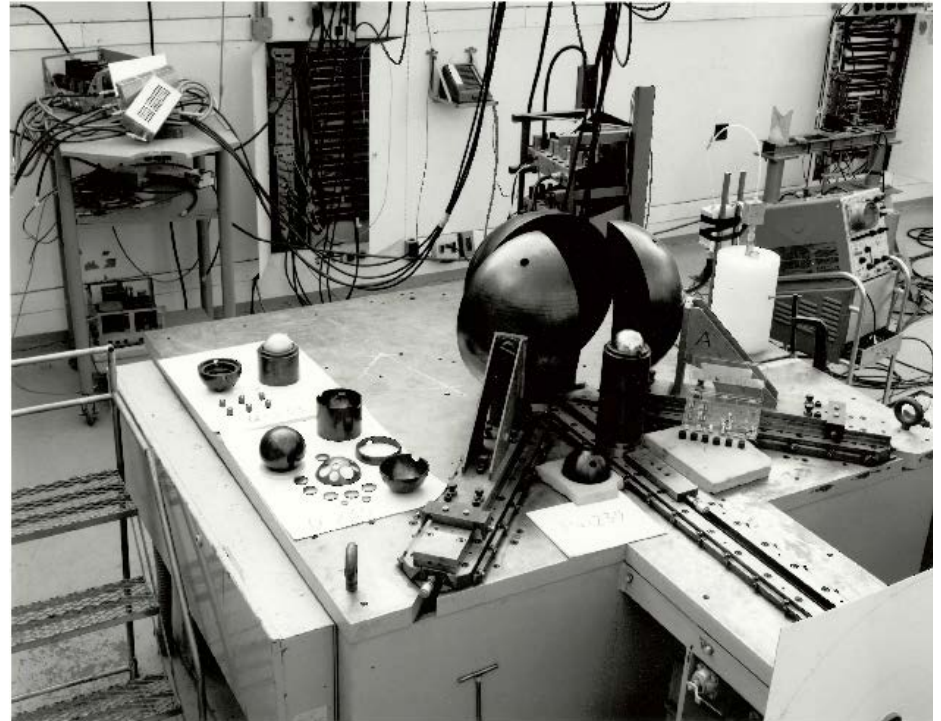


Delta Reactivity (cents) for All Considered Replacement Measurements



Summary

- Knowledge gaps exist on criticality behavior of plutonium as it ages
- An integral experiment has been designed to measure the reactivity worth of new and 50+ year old plutonium
- Experiment based on proven concept with Np-237 in Flattop



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